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Non-equilibrium spin accumulation in ferromagnetic single-electron transistors

Arne Brataas[1,2], Yu. V. Nazarov[1], J. Inoue[1,3], and Gerrit E.W. Bauer[1]

[1] Delft University of Technology, Laboratory of Applied Physics and Delft Institute of Microelectronics and Submicrontechnology (DIMES), 2628 CJ Delft, The Netherlands

[2] Philips Research Laboratories, Prof. Holstlaan 4, 5656 AA Eindhoven, The Netherlands

[3] Nagoya University, Department of Applied Physics, Nagoya, Aichi 46401, Japan

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Abstract. We study transport in ferromagnetic single-electron transistors. The non-equilibrium spin accumulation on the island caused by a finite current through the system is described by a generalized theory of the Coulomb blockade. It enhances the tunnel magnetoresistance and has a drastic effect on the time-dependent transport properties. A transient decay of the spin accumulation may reverse the electric current on time scales of the order of the spin-flip relaxation time. This can be used as an experimental signature of the non-equilibrium spin accumulation.

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1 Introduction

Single-electron tunneling has been an active area of research during the last decade (for a review see Ref. [1]). In a double tunnel junction system, the Coulomb blockade effect is pronounced when the charging energy of the island is larger than the thermal energy, $e^2/2C > k_BT$ (C is the capacitance of the island), and the tunnel resistances are larger than the quantum resistance $R > R_K = h/e^2$. So far most of the research on the double tunnel systems has been in systems using metals (normal or superconducting) or semiconductor hetero-structures [1,2]. The so-called orthodox theory of single-electron tunneling [1] has been very successful in explaining experiments where the

Send offprint requests to: Arne Brataas

Correspondence to: arneb@duttnto.tn.tudelft.nl

single-particle energy separation is smaller than the thermal energy.

Quite unrelated, the giant magnetoresistance in magnetic multilayers and the spin-tunneling magnetoresistance in ferromagnet-insulator-ferromagnet systems have attracted great interest [3,4]. In a magnetic multilayer or in a trilayer "spin-valve" structure the magnetization of the adjacent magnetic layers may vary. When the layers are antiparallel, an external magnetic field can align the magnetization of the ferromagnetic layers. The observed decrease in the resistance is a result of spin-dependent scattering in the materials.

In recent experiments Ono et al. studied the properties of ferromagnetic single-electron transistors (FSETs) [5,6]. This is a new and interesting system in which both the charging aspect due to the Coulomb energy of the excess charge on the island and the magnetoresistance due to the spin-dependent tunneling rates are of importance. Ono et al. found an enhanced magnetoresistance in the Coulomb Blockade regime and a monotonic phase shift of the Coulomb oscillations induced by the magnetic field. The enhancement of the magnetic valve effect in the Coulomb blockade regime has been ascribed to co-tunneling, which is of a higher order in the tunneling resistances and so the difference in the spin-dependent tunneling between the antiparallel and the parallel configuration is larger [6,7]. The magneto-Coulomb oscillations can be explained in terms of changes in the free energy of the island electrode and the leads in the presence of an external magnetic field [8]. Coulomb charging effects have also been seen in discontinuous multilayers [9] and in small cobalt clusters [10].

A generalization of the orthodox theory to describe a FSET by introducing spin-dependent tunneling rates has been presented by Barnas and Fert [11] and Majumdar and Hershfield [12]. However they neglected the effects of a non-equilibrium spin accumulation on the island caused by the spin-dependent tunneling rates, which can have a drastic effect on the transport properties of the ferromagnetic single-electron transistor.

In a ferromagnetic single-electron transistor, the transition rates for tunneling into or out of the island depend on the electron spin. An electric current through the system therefore implies a spin current out of or into the island $(\partial s/\partial t)_{\mathrm{tr}}$. This creates a non-equilibrium excess spin on the island. The excess spin s decays by spin-flip relaxation so that $s = (\partial s/\partial t)_{\mathrm{tr}}\tau_{\mathrm{sf}}$, where τ_{sf} is the spin-flip relaxation time. The non-equilibrium spin accumulation on the island is equivalent to a non-equilibrium chemical potential difference $\Delta\mu$ between the spin-up and the spin-down states. In the case of a normal metal island this chemical potential difference can be evaluated as $\Delta\mu = s\delta$, where δ is the single-particle energy spacing at the Fermi energy (inverse density of states). The chemical potential difference modifies the transport properties. The non-equilibrium spin accumulation is important for the transport properties of the FSET when the non-equilibrium chemical potential difference $\Delta\mu$ is of the same order as the Coulomb charging energy $E_c = e^2/2C$. The spin current is of the same order as the electric cur-

rent, $e(\partial s/\partial t)_{\text{tr}} \sim I$. We are interested in voltages of the order of the Coulomb gap, so we have $I \sim GE_c/e$, where G is the typical junction conductance. From these crude considerations we deduce that the non-equilibrium spin accumulation is important when the spin-flip relaxation time and/or the single-particle energy separation are sufficiently large, i.e. when

$$\tau_{\text{sf}}\delta/h > R/R_K, \tag{1}$$

irrespective of E_c.

The spin-flip relaxation time is crucial for the observation of the non-equilibrium spin accumulation. The spin-flip scattering in clean metals at low temperatures is dominated by the spin-orbit scattering, $\tau_{\text{sf}} \approx \tau_{\text{SO}}$. Since spin-orbit coupling is a relativisitic effect, it increases with the atomic number Z of the metal. The single-particle energy spacing on the island increases with decreasing number of atoms N on the island as $\delta \sim E_F/N$, where E_F is the Fermi energy. Thus the size of the island must be small for heavy elements in order to observe big effects of the spin-accumulation. Let us first estimate the spin-flip relaxation time in an island with an ideal clean surface, where the spin-flip scattering at the boundary of the island can be disregarded. The spin-flip relaxation time in single-crystal aluminium is ($\tau_{\text{sf}} \sim 10^{-8}s$ at $T = 4.3K$ [13] ($\tau_{\text{sf}} \sim 10^{-10}s$ in polycrystalline aluminium [14]) and $\tau_{\text{sf}} \sim 10^{-11}s$ for gold[15]. The Fermi energy is $E_F \sim 10eV$, and the spin accumulation may therefore be expected to play a significant role in an Al island with less than 10^8 atoms (10^6 atoms in polycrsalline aluminium). In the limit $\tau_{\text{sf}} \to 0$ or for a large island, our results reduce to the models in Refs. [11,12], where the spin was assumed to be equilibrated.

Spin-accumulation is important in small systems also when the boundary roughness gives a dominant contribution to the spin-flip scattering. In such a case, the spin-orbit relaxation time is $\tau_{\text{SO}} = f\tau$, where f is the ratio of non spin-flip to spin-flip scattering rates and τ is the momentum relaxation time. For a small system with an ideally rough surface $\tau \sim L/v_F$, where L is the size of the system and v_F is the Fermi velocity. Abrikosov and Gor'kov calculated [16] $f = (\alpha Z)^{-4}$, where $\alpha = e^2/(\hbar c) \simeq 1/137$ is the fine-strucure constant, in rough agreement with experiments [17]. Using $L \sim aN^{1/3}$, where $a \sim 1/k_F$ is the lattice constant $\tau \sim N^{1/3}/(\hbar E_F)$ and the relation (1) reads for small particles with rough boundaries:

$$N < \left(f\frac{R_K}{R}\right)^{3/2}. \tag{2}$$

Assuming that the tunnel resistance is of the order of the quantum resistance (e.g. in the measurement by Ono et al. [6] $R/R_K \sim 1$), and using estimates for the ratio of non spin-flip to spin-flip scattering rates from the measurements in Ref. [17], we expect to see effects of a non-equilibrium spin accumulation in Cu ($\log f \sim 2.1 - 2.4$) when $N < 10^3$, in Al ($\log f \sim 2.8 - 3.6$) when $N < 10^4 - 10^5$, in Na ($\log f \sim 4.2 - 5.3$) when $N < 10^6 - 10^9$ and in Li ($\log f \sim 4.9 - 7$) when $N < 10^7 - 10^{10}$. For smooth boundaries, these numbers should be correspondingly larger. For our purposes, the study of effects of a non-equilibrium spin accumulation, one should choose a light metal with a long spin-orbit relaxation time (a large parameter f).

Note that the orthodox model is still valid for the small clusters described above. For a small island, the capacitance scales like $C \sim aN^{1/3}$. The ratio between the single-particle energy spacing and the Coulomb charging energy is therefore $\delta/E_C \sim \left(E_F a/e^2\right) N^{-2/3}$. The prefactor is $\left(E_F a/e^2\right) \sim 1$, and thus the single-particle spacing is much smaller than the Coulomb charging energy. Our generalized orthodox model is therefore valid when the temperature is larger than the single-particle energy spacing. For lower temperatures, the single-particle energy spacing will also appear in the current-voltage characteristics.

"Modern" metals, like arm-chair nanotubes [18] or (magnetic) semiconductor heterostructures [19] can also be interesting as island materials. The first because of a possible huge spin-flip relaxation time ($Z = 6$ for carbon) and the latter since islands containing a small number of electrons can be created by depletion of the two-dimensional electron gas [1]. However, in these systems quantum size effects start to play a role which have our attention.

In small tunneling systems where Eq. (1) is satisfied, the spin-flip relaxation time is longer than the charge relaxation time RC. This can be seen from Eq. (1), $\tau_{\mathrm{sf}} > (2E_c/\delta)RC$, and noting that the charging energy is larger than the single-particle energy spacing for all but in few-electron systems. Hence the long-time response of the system is dominated by the spin dynamics. We will demonstrate that this feature can be used as an experimental signature of the non-equilibrium spin accumulation.

The paper is organized in the following way. In the next section 2 we model the transport in the ferromagnetic single-electron transistor by generalizing the orthodox theory. The simple case of a halfmetallic ferromagnet-normal metal-halfmetallic Ferromagnetic (HF/N/HF) FSET can be treated analytically and is considered in section 3. The transport properties in the general case are investigated numerically in section 4. Finally our conclusions are presented in section 5. Selected results of the present work have been presented in Ref. [20].

2 Model

We consider the situation shown in Fig. 1. A (ferromagnetic or normal) metal island is attached to two (ferromagnetic or normal) leads by two tunnel junctions. There is an applied source-drain voltage V between the right and the left reservoir and a gate voltage source coupled capacitively to the island. We assume collinear magnetization in the leads and the island. The direction of the magnetizations in the island and the right lead can be parallel or antiparallel to the direction of the magnetization in the left reservoir as depicted by the broken arrows in the Fig. (1). When the coercive fields of the magnets are different different configurations of the magnetizations can be realized by sweeping an external magnetic field. We ignore for simplicity the complications due to the possible appearance of a magnetic domain structure in the ferromagnets. The tunnel junctions have a capacitance C_i and spin-dependent conductances $G_{i\sigma}$, where $i = 1, 2$ denotes the first and the second junction and σ denotes up (+) or down (−) spin electrons. The spin-dependent conductance of the first junction, $G_{1\sigma}$, is proportional to the spin-dependent

density of states in the left reservoir at the Fermi level $N_{\mathrm{L}\sigma}(0)$, the island at the Fermi level $N_{\mathrm{I}\sigma}(0)$ and the angular averaged spin-dependent tunneling probability at the Fermi level, $G_{1\sigma} = 2\pi e^2 N_{L\sigma}(0) N_{I\sigma}(0) T_{1\sigma}(0)/\hbar$ [21]. In a ferromagnet the tunneling probability is significantly different for the s- and d-electrons. The density of states and the tunneling probabilities should therefore be regarded as phenomenological parameters which are to be constant in an interval of the order of the applied source-drain voltage at the Fermi surface. This assumption has been found to be valid if the applied source-drain voltage is lower than 100 mV [11]. The expression for the spin-dependent conductance of the second junction is similar. We take the gate capacitance C_g to be small compared to the junction capacitances C_1 and C_2 and other impedances are disregarded. The energy difference associated with the tunneling of one electron into the island through junction i is [1]

$$E_i(V,q) = \kappa_i eV + \frac{e(q - e/2)}{C_1 + C_2},$$

where $q = -ne + q_0$ is the charge of the island, the total capacitance is $1/C = 1/C_1 + 1/C_2$ and $\kappa_i = C/C_i$. The number of excess electrons on the island is n. The offset charge q_0 is controlled by the gate voltage coupled to the island by the gate capacitance, $q_0 = C_g V_g$.

At a finite current through the island, the energy diagram in Fig. 2 should be considered. Here we show the equilibrium chemical potentials in the left and the right reservoir and the non-equilibrium chemical potentials for the spin-up and the spin-down electrons in the island. It is assumed that the energy relaxation in the island is much faster than the time between the tunneling events $\sim RC$, so that the distributions of the energy levels for the different spins are described by Fermi functions. The non-equilibrium chemical potentials in the island are spin-split by $\Delta\mu$ due to the spin accumulation. The non-equilibrium chemical potential difference causes novel phenomena on the single-electron transistor to be discussed below.

The tunnel conductances are taken to be much smaller than the quantum conductance $G_{i\sigma} \ll G_K = 1/R_K$. Thus we disregard co-tunneling [7] and calculate the current-voltage characteristics in the system by using the semiclassical master equation. The transition rates can be found from Fermi's Golden Rule [1]. The spin-dependent tunneling rates are

$$\begin{aligned}
\overrightarrow{\Gamma^{1\sigma}}_{n+1,n} &= \frac{1}{e^2} G_{1\sigma} F(E_1(V,q) - \sigma\Delta\mu/2), \qquad (3)\\
\overleftarrow{\Gamma^{1\sigma}}_{n,n+1} &= \frac{1}{e^2} G_{1\sigma} F(-E_1(V,q) + \sigma\Delta\mu/2),\\
\overleftarrow{\Gamma^{2\sigma}}_{n+1,n} &= \frac{1}{e^2} G_{2\sigma} F(E_2(-V,q) - \sigma\Delta\mu/2),\\
\overrightarrow{\Gamma^{2\sigma}}_{n,n+1} &= \frac{1}{e^2} G_{2\sigma} F(-E_2(-V,q) + \sigma\Delta\mu/2).
\end{aligned}$$

$\overrightarrow{\Gamma^{1\sigma}}_{n+1,n}$ denotes transition from the left reservoir to the island, so that the number of electrons on the island is changed from n to $n+1$, etc. and

$$F(E) = \frac{E}{1 - \exp(-E/k_B T)},$$

where $k_B T$ is the thermal energy. We also define the total forward rates $\overrightarrow{\Gamma^{i}} = \overrightarrow{\Gamma^{i\uparrow}} + \overrightarrow{\Gamma^{i\downarrow}}$, where $i = 1, 2$, and analogous backward rates. The combined rate for an increase of the number of excess electrons on the island is $\Gamma_{n+1,n} = \overrightarrow{\Gamma^{1}}_{n+1,n} + \overleftarrow{\Gamma^{2}}_{n+1,n}$ and analogous $\Gamma_{n,n+1} = \overleftarrow{\Gamma^{1}}_{n,n+1} + \overrightarrow{\Gamma^{2}}_{n,n+1}$. The master equation which determines the prob-

ability p_n of having n excess electrons on the island is

$$\frac{dp_n}{dt} = -p_n \left(\Gamma_{n-1,n} + \Gamma_{n+1,n} \right) + p_{n+1}\Gamma_{n,n+1} + p_{n-1}\Gamma_{n,n-1}. \tag{4}$$

In the stationary state $dp_n/dt = 0$ and the detailed balance symmetry gives $\Gamma_{n+1,n}p_n = \Gamma_{n,n+1}p_{n+1}$. The current through the first junction is $I_1 = (I_1^{\uparrow} + I_1^{\downarrow})$, where the current of electrons with spin σ is

$$I_1^{\sigma} = e \sum_n p_n \left(\overrightarrow{\Gamma^{1\sigma}}_{n+1,n} - \overleftarrow{\Gamma^{1\sigma}}_{n-1,n} \right).$$

In the second junction the current is $I_2 = (I_2^{\uparrow} + I_2^{\downarrow})$,

$$I_2^{\sigma} = e \sum_n p_n \left(\overrightarrow{\Gamma^{2\sigma}}_{n-1,n} - \overleftarrow{\Gamma^{2\sigma}}_{n+1,n} \right).$$

The transport of spins into the island is determined by

$$\left(\frac{ds}{dt}\right)_{\rm tr} = \left(\frac{d\left(N_\uparrow - N_\downarrow\right)}{dt}\right)^{\rm in} - \left(\frac{d\left(N_\uparrow - N_\downarrow\right)}{dt}\right)^{\rm out} = (I_1^{\uparrow} - I_2^{\uparrow} - I_1^{\downarrow} + I_2^{\downarrow})/e. \tag{5}$$

In the stationary situation we can use the current conservation $I_1^{\uparrow} + I_1^{\downarrow} = I_2^{\uparrow} + I_2^{\downarrow}$ and find

$$\left(\frac{ds}{dt}\right)_{\rm tr}^{\rm st.} = 2(I_1^{\uparrow} - I_2^{\uparrow})/e = 2(I_2^{\downarrow} - I_1^{\downarrow})/e. \tag{6}$$

The spin balance is

$$\frac{ds}{dt} = \left(\frac{ds}{dt}\right)_{\rm tr} + \left(\frac{ds}{dt}\right)_{\rm rel}, \tag{7}$$

where $(ds/dt)_{\rm rel}$ is the spin-flip relaxation rate. In equilibrium there are s_0 spins on the island (for a normal metal island $s_0 = 0$). The chemical potential is μ_0. The non-equilibrium spin-dependent chemical potentials are $\mu_\uparrow = \mu_0 + \delta\mu + \Delta\mu/2$ and $\mu_\downarrow = \mu_0 + \delta\mu - \Delta\mu/2$. The total number of spins on the island is $s = s_0 + [(\delta\mu + \Delta\mu/2)\delta_\uparrow^{-1} - (\delta\mu - \Delta\mu/2)\delta_\downarrow^{-1}]$, where δ_σ^{-1} is the spin-dependent density of states. The spin-flip relaxation rate is

$$\left(\frac{ds}{dt}\right)_{\rm rel} = -\frac{s - \overline{s}}{\tau_{\rm sf}} = -\frac{\Delta\mu}{\tau_{\rm sf}\delta}. \tag{8}$$

In the limit of fast spin-flip relaxation ($\tau_{\rm sf} \to \infty$), the number of spins on the island is $\overline{s} = s_0 + \delta\mu(\delta_\uparrow^{-1} - \delta_\downarrow^{-1})$. Eq. (7) determines the non-equilibrium chemical potential shift $\Delta\mu$. Here $\tau_{\rm sf}$ is the spin-flip relaxation time in the island and $\delta^{-1} = (\delta_\uparrow^{-1} + \delta_\downarrow^{-1})/2$ is the average density of states for spin up and spin down electrons at the Fermi level in the island (the inverse single-particle energy spacing). Eq. (8) effectively includes many-body effects. The excess spin is related to the energy difference between spin-up and spin-down states by $s - \overline{s} = \chi_s \Delta\mu/\mu_B^2$, where χ_s is the spin susceptibility (for noninteracting electrons $\chi_s = \mu_B^2\delta^{-1}$). In the stationary state ($dp_n/dt = 0$, $ds/dt = 0$), the spin balance (7) can be written as

$$I_s = e\left(\frac{ds}{dt}\right)_{\rm tr} = G_s 2\Delta\mu/e, \tag{9}$$

where the spin-flip conductance is defined by

$$G_s = \frac{e^2}{2\tau_s\delta}. \tag{10}$$

From the equations for the tunneling rates (3) and the spin balance (7), it is not directly obvious that there is only one unique solution for the non-equilibrium chemical potential difference $\Delta\mu$ for a given set of parameters (temperature, gate voltage, source-drain voltage, capacitances and conductances). If Eq. (7) would be fulfilled for multiple solutions $\Delta\mu$ for a given set of parameters, the current-voltage characteristics has a hysteretic behavior, with a solution $\Delta\mu$ depending on the history. However, by extensive analytical and numerical studies with many dif-

ferent parameters to be presented below we always found only one unique solution of Eq. (7).

In this orthodox model the problem can be mapped on the equivalent circuit in Fig. 3 by introducing a "spin-flip capacitance" $C_s \equiv e^2/2\delta$, so that

$$(es)/2 = C_s(\Delta\mu/e),\ \Delta\mu/s = e^2/(2C_s) = \delta.$$

This "charging energy" of the spin-flip capacitance is thus simply the single-particle energy cost of a spin-flip, δ, or more generally, the inverse of the magnetic susceptibility μ_B^2/χ_s.

Let us first discuss the situations in which $\Delta\mu$ vanishes identically in the stationary state. From the expressions for the spin transport (5), the current conservation through the two junctions, $I = I_1 = I_2$, and the spin-dependent rates (3), we see that when $\Delta\mu = 0$ the spin-dependent currents are related to the total current by

$$I_i^{\uparrow} = \frac{1}{1 + G_{i\downarrow}/G_{i\uparrow}} I,$$
$$I_i^{\downarrow} = \frac{1}{1 + G_{i\uparrow}/G_{i\downarrow}} I,$$

where $i = 1, 2$,

$$I_s = 2I\left(\frac{1}{1 + G_{1\downarrow}/G_{1\uparrow}} - \frac{1}{1 + G_{2\downarrow}/G_{2\uparrow}}\right).$$

From this relation we can make the following observations. First, we see that in the symmetric case, $G_{1\uparrow}/G_{1\downarrow} = G_{2\uparrow}/G_{2\downarrow}$, the spin-current is zero for $\Delta\mu = 0$ and therefore $\Delta\mu = 0$ is a solution of the spin balance equation (7) for any source-drain voltage V, gate voltage V_g and temperature T, i.e. *there is a solution with zero non-equilibrium chemical potential shift*. This symmetry is expected for a structure of the type (Ferromagnet A- Ferromagnet B - Ferromagnet A) with magnetization configuration $\uparrow\uparrow\uparrow$ or $\uparrow\downarrow\uparrow$, which is the case for the device measured in Refs. [5, 6]. Thus in this case our theory reduces to those in Refs. [11,12], where the spins were assumed to be equilibrated. Second, we find that for a general system in the Coulomb blockade regime the current is zero, $I = 0$, and hence a vanishing non-equilibrium chemical potential difference $\Delta\mu = 0$ is a solution in the Coulomb blockade regime. This is physically reasonable since it means that there is no spin accumulation on the island when there is no current flowing through the junctions. The Coulomb gap in the current-voltage characteristics is not modified by the spin accumulation.

3 Analytical results for a halfmetallic ferromagnet/normal metal/ halfmetallic ferromagnet FSET

We will now discuss an idealized case for which an analytic expression for the current-voltage can be derived at zero temperature. Let us assume that the leads are halfmetallic ferromagnets and the island is a normal metal. The density of states at the Fermi energy vanishes for the minority spins in a halfmetallic ferromagnet. The capacitances are taken to be symmetric, $C_1 = C_2 = C$, and the gate voltage is tuned so that the offset charge is zero $q_0 = 0$. When the direction of the magnetizations in the ferromagnetic leads are antiparallel the tunnel conductances are

$$G_{1\uparrow}^{\mathrm{AP}} = G_1, G_{1\downarrow}^{\mathrm{AP}} = 0, G_{2\uparrow}^{\mathrm{AP}} = 0, G_{2\downarrow}^{\mathrm{AP}} = G_2. \quad (11)$$

The conductances $G_{1\downarrow}^{\mathrm{AP}}$ and $G_{2\uparrow}^{\mathrm{AP}}$ are zero, since there are no spin-down states at the Fermi surface in the left reservoir and there are no spin-up states at the Fermi surface in the right reservoir. The electric current is $I = I_1^{\uparrow}$ and the spin-current is

$$e\left(\frac{ds}{dt}\right)_{\mathrm{tr}} = 2(I_1^{\uparrow} - I_2^{\uparrow}) = 2I.$$

The spin-current is directly proportional to the current for any temperature T and source-drain voltage V. In the absence of spin-flip relaxation, $\tau_{\mathrm{sf}} \to \infty$, the current through the system vanishes, because an electron cannot propagate from the left reservoir to the right reservoir without spin-flip. The master equation can be solved exactly at zero temperature for specific voltages V_m^{AP} [22]:

$$\frac{1}{2}\left(eV_m^{\mathrm{AP}} - \Delta\mu\right) = E_c\left(m + \frac{1}{2}\right),$$

where $E_c = e^2/2C$ is the Coulomb charging energy and m is an integral number. The non-vanishing rates are

$$\begin{aligned}
\overrightarrow{\Gamma^{1\uparrow}}_{n+1,n} &= \frac{1}{e^2}G_1 E_c(m-n)\Theta(m-n), \qquad (12)\\
\overleftarrow{\Gamma^{1\uparrow}}_{n,n+1} &= -\frac{1}{e^2}G_1 E_c(m-n)\Theta(n-m),\\
\overleftarrow{\Gamma^{2\downarrow}}_{n+1,n} &= -\frac{1}{e^2}G_2 E_c(m+n+1)\Theta(-m-n-1),\\
\overrightarrow{\Gamma^{2\downarrow}}_{n,n+1} &= \frac{1}{e^2}G_2 E_c(m+n+1)\Theta(m+n+1),
\end{aligned}$$

where $\Theta(x)$ is the Heaviside function. The probability of finding n electrons on the island is $(n \leq m)$ [22]

$$p_n(m) = \left(\frac{G_1}{G_2}\right)^n \frac{(m!)^2}{(m-|n|)!(m+|n|)!} p_0(m)$$

and $p_0(m)$ can be found from the normalization condition $\sum_n p_n(m) = 1$ [22]. The current is

$$I = G_{1,2}\left(V_m^{\mathrm{AP}} - \frac{\Delta\mu + E_c}{e}\right),$$

where we have defined the conductance for the two junctions in series, $G_{1,2} = G_1G_2/(G_1+G_2)$. The non-equilibrium chemical potential difference is determined by Eq. (9) which is simplified to $I = I_s/2 = G_s\Delta\mu/e$ as

$$\Delta\mu = \frac{G_{1,2}}{G_s + G_{1,2}}\left(eV_m^{\mathrm{AP}} - E_c\right).$$

The current in the system can then be found to be

$$I^{\mathrm{AP}} = \frac{G_sG_{1,2}}{G_s + G_{1,2}}\left(V_m - \frac{E_c}{e}\right) \qquad (13)$$

at specific voltages determined by

$$eV_m^{\mathrm{AP}} = E_c\left(2m\frac{G_s + G_{1,2}}{G_s} + 1\right).$$

We see from the current (13) that the Coulomb blockade threshold is unchanged as expected. On entering the island from one of the leads the electrons must flip their spins in order to be able to tunnel through the other lead. The total conductance of the system is thus given by the three resistances in series, $1/G_{\mathrm{tot}}^{\mathrm{AP}} = 1/G_s + 1/G_1 + 1/G_2$ as can be seen from the equivalent circuit in Fig. 3. No instabilities (i.e. multiple solutions of the non-equilibrium chemical potential difference $\Delta\mu$ for a given set of parameters) exist.

For a parallel orientation of the magnetizations in the two leads, where $\Delta\mu = 0$, the current-voltage characteristics is [22]

$$I^{\mathrm{P}} = G_{1,2}(V_m^{\mathrm{P}} - \frac{E_c}{e}), \qquad (14)$$

where $eV_m^{\mathrm{P}} = E_c(2m+1)$. Note that the specific voltages for which we have determined the current differ for the parallel and the antiparallel situation. The junction magnetoresistance is determined by

$$\mathrm{JMR} = (I^{\mathrm{P}} - I^{\mathrm{AP}})/I^{\mathrm{P}} \qquad (15)$$

at the same voltages, which we cannot calculate exactly. At high source-drain voltages at which the Coulomb charging effects can be disregarded we see from Eq. (13) and Eq. (14) and Fig. 3 that the magnetoresistance for the system is

$$\mathrm{JMR} = \frac{G_{1,2}}{G_s + G_{1,2}}.$$

In the limit of no spin-flip relaxation, $\tau_{\mathrm{sf}} \to \infty$ ($G_s \to 0$), the junction magnetoresistance is 100 % since the current vanishes in the antiparallel configuration and in the limit of perfect spin-flip relaxation, $\tau_{\mathrm{sf}} \to 0$ ($G_s \to \infty$), the junction magnetoresistance vanishes.

4 General configurations: Numerical results and discussions

In the general situation with arbitrary junction conductances and capacitances and at finite temperatures, the spin-current and the electric current in the system have to be calculated numerically. In experiments the tunnel conductances of the two junctions depend strongly on the thickness of the oxide tunnel barriers. We therefore present numerical results for a variety of possible realizations of the junction parameters.

We choose symmetric capacitances $C_1 = C_2 = C$ in our calculations. The important energy scale is then the Coulomb energy $E_c = e^2/2C$ and we scale the other relevant energies by this energy. The spin-dependent junction conductances in the two junctions are described in units of a typical junction conductance G, so that the electric current and the spin-current are calculated in units of the typical current $Ge/2C$.

Let us first consider the DC transport properties. In a calculation of the current through the system for a given source-drain voltage V, we must first determine the non-equilibrium chemical potential difference $\Delta\mu$ by the spin balance on the island (9). The solution of this equation is given by the intersection of the spin-current into or out of the island $I_s(\Delta\mu)$ and the straight line $G_s 2\Delta\mu/e$ which describes the spin-flip relaxation in the island, where the spin-flip conductance G_s was defined in Eq. (10).

As we have pointed out in the previous chapter, the non-equilibrium spin does not modify the Coulomb gap. However it does affect the current when the source-drain voltage is larger than the Coulomb gap. We show in Fig. 4 the current as a function of the source-drain voltage V for a system consisting of $G_{1\uparrow}/G = 0.3$, $G_{1\downarrow}/G = 0.1$, $G_{2\uparrow}/G = 3$, $G_{2\downarrow}/G = 6$, $C_1 = C_2 = C$, $q_0 = 0$ and the thermal energy $k_B T = 0.02 E_c$. The upper curve shows the current when spin relaxation is fast $G_s/G = 1000$, and the lower curve shows the current in the case of slow spin relaxation, $G_s/G = 0$. The Coulomb blockade threshold for low source-drain voltages is clearly seen and the steplike structure for higher voltages called the Coulomb staircase resulting from the discrete charging of the island [1]. The Coulomb blockade and the Coulomb staircase are smeared by the thermal fluctuations. Since the conductances of the tunnel junctions are spin-dependent, the spin-flip relaxation on the island is important for the current. The current increases with increasing spin-flip con-

ductance G_s (decreasing spin-flip relaxation time τ_{sf}). In the case of fast spin-flip relaxation ($G_s/G = 1000$), the spin-accumulation is vanishing small, and the steps occur at $eV = (2n+1)e^2/2C$, where $n = 0, 1, 2, ...$. For $G_s/G = 0$, there is a spin- accumulation when $eV > e^2/2C$, so that the relative energy difference associated with the tunneling of one electron onto the island $E_i(V, q) - \sigma\Delta\mu/2$ is spin-dependent. Hence, the spin-degenerate staircase at $eV = (2n+1)e^2/2C$ for $n = 1, 2, 3...$ splits into two peaks, where the splitting is proportional to the spin-accumulation. This can be seen in the lower curve in Fig. 4, where the second staircase ($2n+1 = 3$) occur at a voltage lower than $3e^2/2C$ [23].

Another typical experiment on the single-electron transistor is to measure the differential conductance in the linear source-drain voltage regime as a function of the gate voltage V_g ($q_0 = V_g C_g$). We show in Fig. 5 the influence of the spin-flip relaxation in the island on the differential conductance. The system is described by $G_{1\uparrow}/G = 3.0$, $G_{1\downarrow}/G = 0.2$, $G_{2\uparrow}/G = 0.1$, $G_{2\downarrow}/G = 2.0$, $C_1 = C_2 = C$ and the thermal energy is $k_B T = 0.1E_c$. The curves show the differential conductance as a function of the gate voltage. The upper curve is calculated for fast spin relaxation $G_s/G = 500$, the mid curve is for intermediate spin relaxation $G_s/G = 0.5$ and the lower curve is for slow spin relaxation $G_s/G = 0.005$. The typical oscillatory dependence of the differential conductance with respect to the gate voltage is seen. In the same way as the current at high voltages increases with increasing spin-flip relaxation in Fig. 4, we see that the differential conductance increases with increasing spin-flip conductance (decreasing spin-flip relaxation time). As usual the peaks in the conductance as a function of the gate voltage increases with decreasing temperature (not shown) [1].

The junction magnetoresistance is the relative difference in the resistance on switching the directions of the magnetizations in the leads from parallel to antiparallel (15). Let us consider the situation where the leads are ferromagnetic and the island is non-magnetic, i.e. a F/N/F junction. The spin-accumulation causes a non-zero magnetoresistance. In the parallel configuration, the conductances are $G_{1\sigma} = G_1(1+\sigma P)/2$ and $G_{2\sigma} = G_2(1+\sigma P)/2$, where P is the polarization of the ferromagnet. We show in Fig. 6 the calculated junction magnetoresistance for $G_1 = G$, $G_2 = 3G$, $V_g = 0$, $k_B T = 0.05E_c$, $C_1 = C_2 = C$ and a polarization $P = 0.3$ in the limit of slow spin-flip relaxation $G_s = 0$ (upper curve), intermediate relaxation $G_s/G = 1$ (mid curve) and fast spin-flip relaxation $G_s = 4G$ (lower curve). At low source-drain bias, we observe the magnetoresistance oscillations already reported in [11,12]. The amplitude of the oscillations decreases with increasing source-drain voltage, thus decreasing importance of the Coulomb charging [11,12]. The period of the oscillations for our system is close to $2E_c$. There is only a small distortion of the shape of the magnetoresistance oscillations with increasing spin-flip relaxation rate in the island. The oscillations in the TMR as a function of the source-drain voltage can be understood as a consequence of the spin-accumulation in the antiparallel configuration. The spin-accumulation increases with increasing current

through the system. We have seen in Fig. 4 that the current has a steplike behavior with a period close to $2E_c$ due to the discrete charging of the island. Hence the spin-accumulation also shows a steplike behavior (not shown) as a function of the source-drain voltage. The magnetoresistance for the F/N/F FSET increases with increasing spin-accumulation and hence shows oscillations with a period $2E_C$. The magnetoresistance is noticeable even when the spin-flip conductance is of the same order as the tunnel conductances in agreement with Eq. (1). Disregarding the Coulomb charging energy, the junction magnetoresistance is

$$\mathrm{JMR} = P^2 \frac{1-\gamma^2}{1-P^2\gamma^2+\alpha^2}, \tag{16}$$

where $\gamma = (G_1 - G_2)/(G_1 + G_2)$ is a measure of the asymmetry of the junction conductances and $\alpha^2 = 4G_s/(G_1 + G_2)$ determines the reduction of the magnetoresistance due to the spin-flip relaxation. At a high source-drain bias, the numerical results agree well with Eq. (16), giving JMR= 6.9% for $G_s/G = 0$, JMR= 3.4% for $G_s/G = 1$ and JMR= 1.4% for $G_s/G = 4$.

So far we have shown how the spin accumulation influences the DC transport properties. Spin accumulation also causes novel features in the AC and transient response of the system. Let us first fix the source-drain voltage at a high bias until the system is stationary and then lower the source-drain voltage. The transient current response in this situation can be *reversed* on time scales of the order of the spin-flip relaxation time, which is an unambigous signature of a non-equilibrium spin. In order to explicitly show this we solved the time-dependent problem by numerically integrating Eq. (4) and Eq. (7). In the upper part of Fig. 7 we show the calculated time-dependent chemical potential difference when the source-drain voltage is suddenly reduced at $t = 0$. The junctions are described by $G_s/G = 0.5$, $G_{1\uparrow}/G = 0.05$, $G_{1\downarrow}/G = 1$, $G_{2\uparrow}/G = 2$, $G_{2\downarrow}/G = 0.01$, $C_1 = C_2 = C$ and the thermal energy is $k_BT = 0.05E_c$. We consider the high voltage case $V = 8E_c$, where the associated stationary electric current is $I = 2.3Ge/2C$ and lower the source-drain voltage to $V = 2E_c$, where the associated stationary electric current is positive, $I = 0.40Ge/2C$. We have used $t_{\mathrm{sf}} = 20RC$, which appears to be a reasonable estimate for junctions with a Coulomb charging energy of $10meV$ and a junction conductance of $R/R_K = 10$ giving a charge relaxation time of $RC = 0.5 \cdot 10^{-12}s$. The chemical potential difference is seen to decay on a time scale much larger than the charge relaxation time. Finally we show in the lower part of Fig. 7 the current through the first junction (full line) and the second junction (dotted line). It is clearly seen that the relaxation of the current is slow on the time scale RC. For time scales up to the order of RC, the currents through the first and the second junction differ due to the depopulation of the charge island. The Coulomb charging effect shows up as the almost constant current on intermediate time-scales.

In order to understand the dynamics it is useful to inspect the device without the Coulomb charging effects, i.e. the capacitances C_1 and C_2 in the equivalent electric circuit in Fig. 3. We set the voltage on the left lead to zero and apply a time dependent potential $V(t)$ to the right

lead. The complex impedance $Z_{\text{spin}}(\omega) = V(\omega)/I(\omega)$ is

$$\frac{1}{Z_{\text{spin}}(\omega)} = \frac{G_1 G_2}{G_1 + G_2} - \frac{G_{1\uparrow}G_{2\downarrow} - G_{1\downarrow}G_{2\uparrow}}{(G_1 + G_2)} \frac{\Delta\mu(\omega)}{eV(\omega)} \quad (17)$$

where

$$\frac{\Delta\mu(\omega)}{eV(\omega)} = \frac{1}{1 + i\omega\tau_{\text{spin}}} \frac{G_{1\uparrow}G_{2\downarrow} - G_{1\downarrow}G_{2\uparrow}}{(G_s + G')(G_1 + G_2)}. \quad (18)$$

Here the spin accumulation time is

$$\tau_{\text{spin}} = \frac{C_s}{G_s + G'}, \quad (19)$$

where $1/G' = 1/(G_{1\uparrow} + G_{2\uparrow}) + 1/(G_{1\downarrow} + G_{2\downarrow})$. From the relations (17) and (18) we see why switching-off the source-drain voltage ($V_f = 0$) reverses the transient current as found in the lower panel in Fig. 7. Without the Coulomb blockade this transient decays on the time scale τ_{spin}. In the limit that the junction conductances are much smaller than the spin conductance, the spin accumulation time (19) reduces to the spin-flip relaxation time, $\tau_{\text{spin}} \approx \tau_{\text{sf}}$. In the opposite limit where the junction conductances are much larger than the spin conductance, the spin accumulation time is $\tau_{\text{spin}} \sim C_s R$. The spin-flip capacitance is much larger than the charge-capacitance C in the regime where the orthodox theory is valid ($\delta \ll E_C$) and thus the spin accumulation time is much larger than the charge-relaxation time. The spin accumulation time in Fig. 7 agree well with the value $t_{\text{spin}} = 0.43 t_{\text{sf}}$ as can be found from the equivalent circuit neglecting the Coulomb blockade. The spin accumulation time deviates from Eq. (19) if the initial or final voltage is less than the Coulomb charging energy [20]. We show in Fig. 8 the average current through the first and the second junction when the final voltage is zero with the same system parameters as above (as used in Fig. 7). In this case the non-equilibrium spin accumulation decays slower since the spins must relax through the spin-flip conductance G_s on the island and the transport through the junctions is suppressed. The spin-accumulation time is then roughly equal to the spin-flip relaxation time [20]. The transient response to switching on the source-drain voltage is similar. If the initial and final voltage are above the Coulomb charging energy, the response time is determined by Eq. (19) otherwise the response time roughly equals the spin-flip relaxation time.

The long time response of the system due to the spin dynamics can also be observed in other AC transport experiments. A fast single-electron transistor has recently been realized[24]. We therefore study the influence of an AC source-drain voltage $V(t) = V_0 + V_1 \cos\omega t$ on the DC current through the system, $\bar{I}(\omega) = \omega/(2\pi) \int_{t_0}^{t_0 + 2\pi/\omega} dt I(t)$, where t_0 is an arbitrary time constant. We show in Fig. 9 the relative change in the DC current

$$R(\omega) = \frac{\bar{I}(\omega = 0) - \bar{I}(\omega)}{\bar{I}(\omega = 0)} \quad (20)$$

as a function of frequency for an applied voltage which fluctuates around the Coulomb blockade threshold voltage, $V(t) = E_c(1 + 0.25\cos\omega t)$. The temperature is $k_B T = 0.05 E_c$, $q_0 = 9$ and $C_1 = C_2$. The system is in the antiparallel configuration so that there is a spin-accumulation on the island, $G_{1\uparrow}/G = 1.5$, $G_{1\downarrow}/G = 0.5$, $G_{2\uparrow}/G = 0.5$,$G_{2\downarrow}/G = 1.5$,$G_s/G = 0.5$ and the spin-flip relaxation time is $\tau_{\text{sf}} = 20RC$. It is seen that the DC current varies most strongly when $\omega\tau_{\text{sf}} \sim 1$, which is a consequence of the spin-dynamics in the system with the characteristic time-scale τ_{sf}. A corresponding calculation with the

same parameters in the *parallel* configuration shows no frequency dependence of $R(\omega)$ around $\omega\tau_{\rm sf} \sim 1$.

5 Conclusions

We have investigated the effect of non-equilibrium spins on the transport properties of a ferromagnetic single-electron transistor. The orthodox theory is generalized to include the spin accumulation on the island. The spin accumulation is more important for small islands with a large energy spacing. The current and the differential conductance increases with increasing spin-flip relaxation rate. The magnetoresistance is enhanced due to the spin accumulation. The non-equilibrium spins on the island can have a drastic effect on the transient transport properties. We have shown that on lowering the source-drain voltage from a high bias to a low bias, a reversed current appears on time scales shorter than the spin-flip relaxation time. The same slow response also appears in other AC transport properties if one or more of the external parameters are time-dependent.

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Fig. 1. The single electron transistor consisting of a (magnetic) island coupled to two (magnetic) reservoirs by tunnel junctions.

Fig. 2. The non-equilibrium chemical potentials in the reservoirs and the island at a finite source-drain voltage and a finite current.

Fig. 3. The equivalent circuit for the current-voltage response of the system.

Fig. 4. The electric current I as a function of the source-drain voltage for different spin relaxation conductances: $G_s/G = 1000$ (upper line) and $G_s/G = 0$ (lower line). The system parameters are $G_{1\uparrow}/G = 0.3$, $G_{1\downarrow}/G = 0.1$, $G_{2\uparrow}/G = 3$, $G_{2\downarrow} = 6$, $T = 0.02 E_C$, $q_0 = 0$, $C_1 = C_2 = 1.0$.

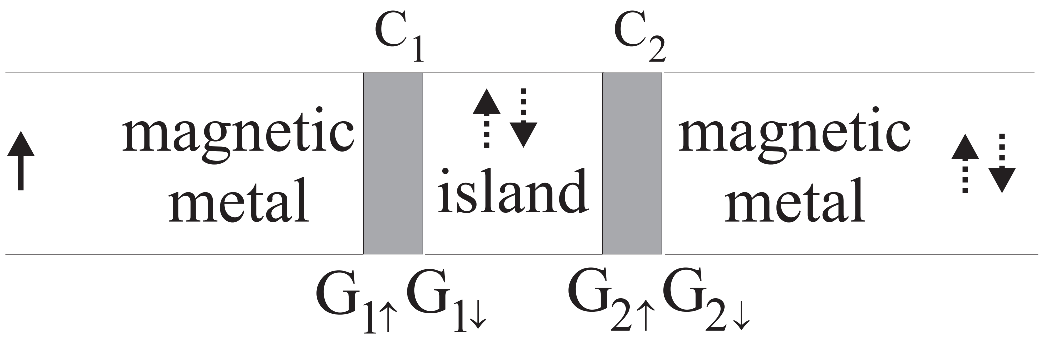
Fig. 5. The conductance dI/dV as a function of the gate-voltage V_g for different spin-relaxation conductances: $G_s/G = 500$ (upper line), $G_s/G = 0.5$ (mid line) and $G_s/G = 0.005$ (lower line). The system parameters are $G_{1\uparrow}/G = 3.0$, $G_{1\downarrow}/G = 0.2$, $G_{2\uparrow}/G = 0.1$, $G_{2\downarrow} = 2.0$, $T = 0.1 E_C$, $q_0 = 0$, $C_1 = C_2 = 1.0$.

Fig. 6. The magnetoresistance as a function of the source-drain voltage in a F/N/F double junction system with P=0.3, $G_{1\uparrow} + G_{1\downarrow} = G$, $G_{2\uparrow} + G_{2\downarrow} = 3G$, $V_g = 0$, $C_1 = C_2 = 1$ and $k_B T = 1.0$. The upper curve is for slow spin-relaxation, $G_s/G = 0$, the mid curve for intermediate spin-flip relaxation $G_s/G = 1$ and the lower curve for faster spin-relaxation, $G_s/G = 4$.

Fig. 7. The current as a function of time. The source-drain voltage is switched from $V = 8E_c$ to $V = 2E_c$ at $t = 0$. The system parameters are $G_s/G = 0.5$, $G_{1\uparrow}/G = 0.05$, $G_{1\downarrow}/G = 1$, $G_{2\uparrow}/G = 2$, $G_{2\downarrow}/G = 0.01$, $k_B T = 0.05 E_C$, $q_0 = 0$ and $C_1 = C_2$.

Fig. 8. The average current through the first and the second junction as a function of time. The source-drain voltage is switched from $V = 8E_c$ to $V = 0$ at $t = 0$. The system parameters are $G_s/G = 0.5$, $G_{1\uparrow}/G = 0.05$, $G_{1\downarrow}/G = 1$, $G_{2\uparrow}/G = 2$, $G_{2\downarrow}/G = 0.01$, $k_BT = 0.05E_c$, $q_0 = 0$ and $C_1 = C_2$

Fig. 9. The time-averaged (DC) current as a function of the frequency of the applied source-drain voltage, $V(t) = E_C(1 + 0.25\cos\omega t)$. The reservoirs are in the antiparallel configuration, $G_{1\uparrow}/G = 1.5$, $G_{1\downarrow}/G = 0.5$, $G_{2\uparrow}/G = 0.5$,$G_{2\downarrow}/G = 1.5$. The other parameters are $k_BT = 0.05E_c$, $q_0 = 9$ and $C_1 = C_2$


C1
C2
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metal
island
magnetic
metal
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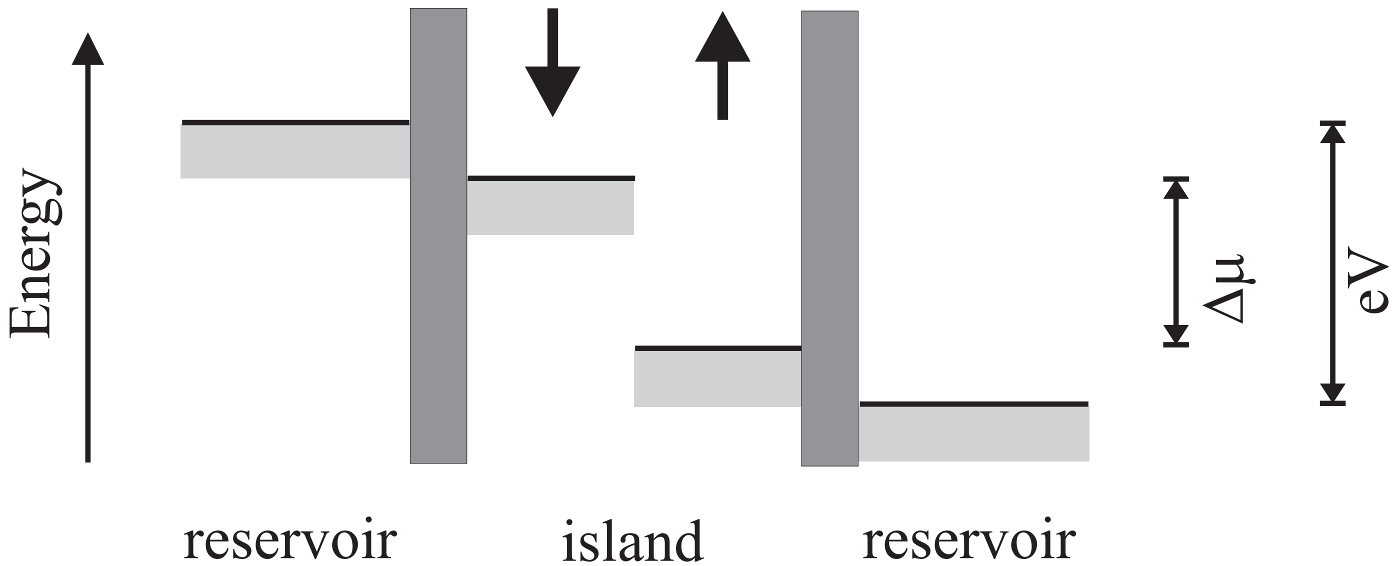
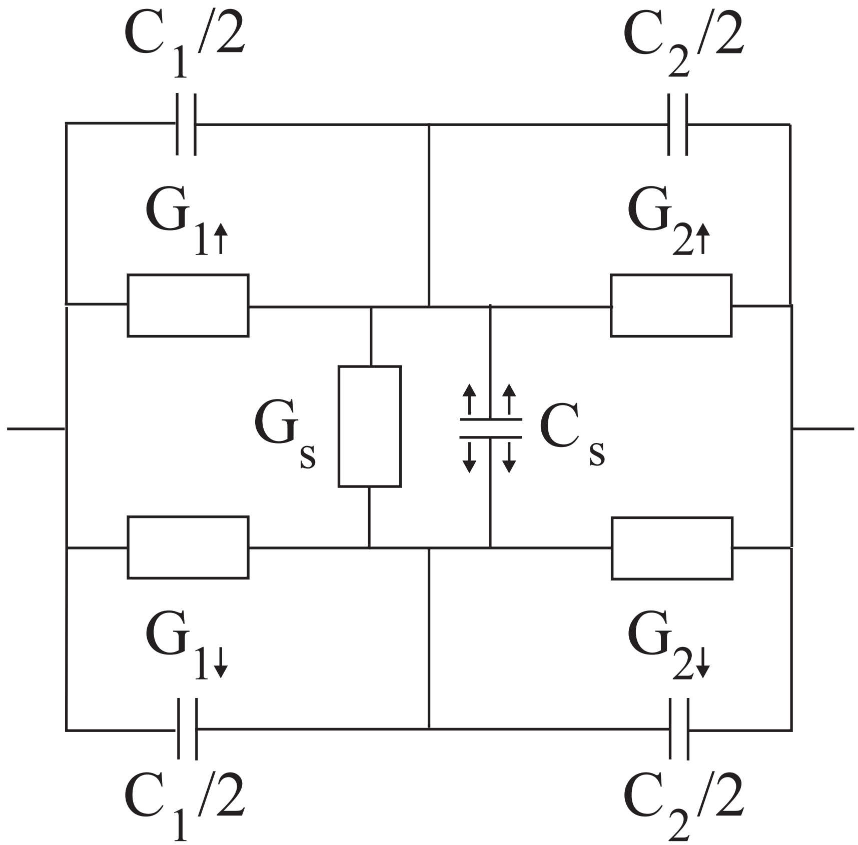
G2↑ G2↓



Energy
Δμ
eV
reservoir
island
reservoir


C 1 /2
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G 1↑
G 2↑
G s
C s
G 1↓
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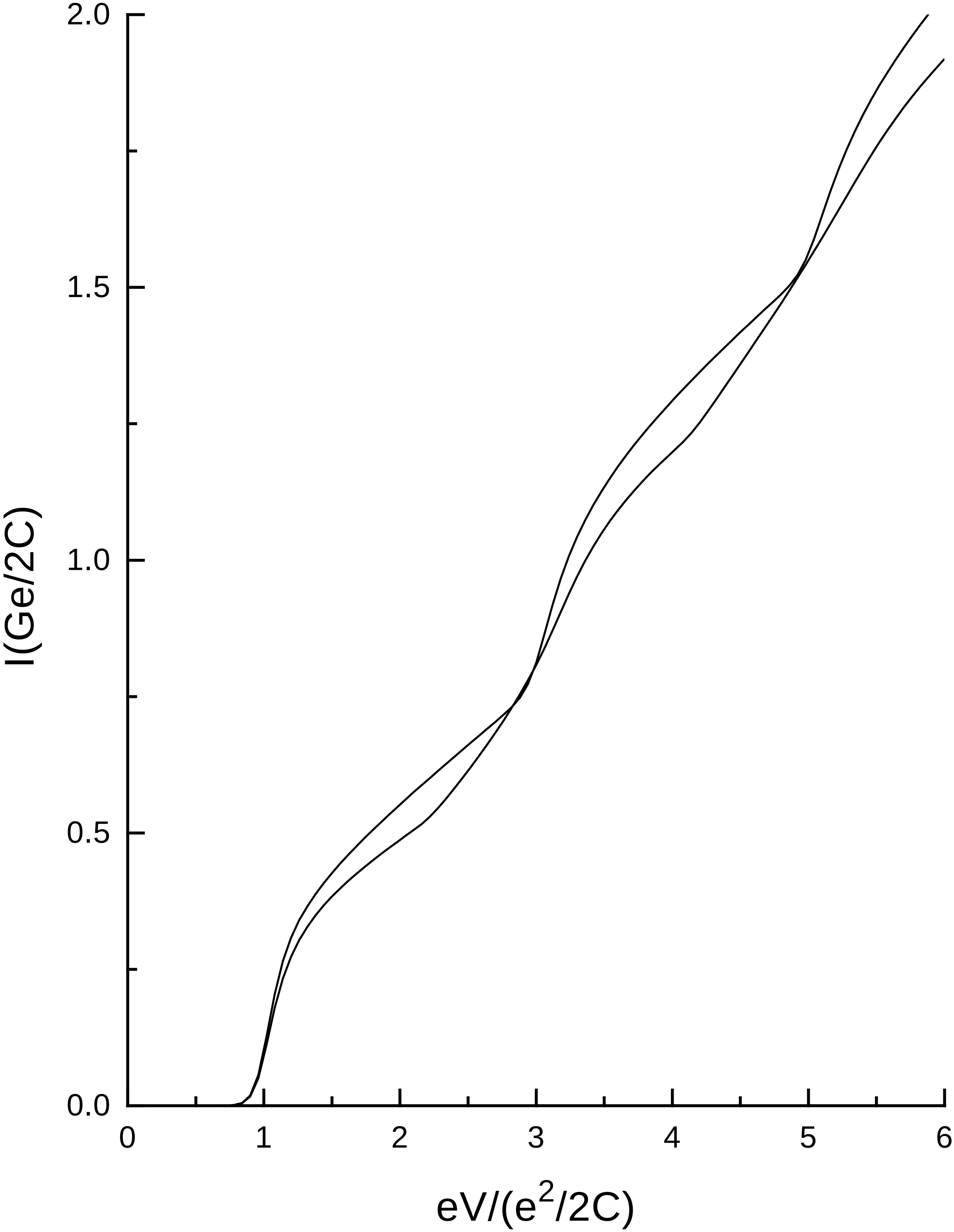
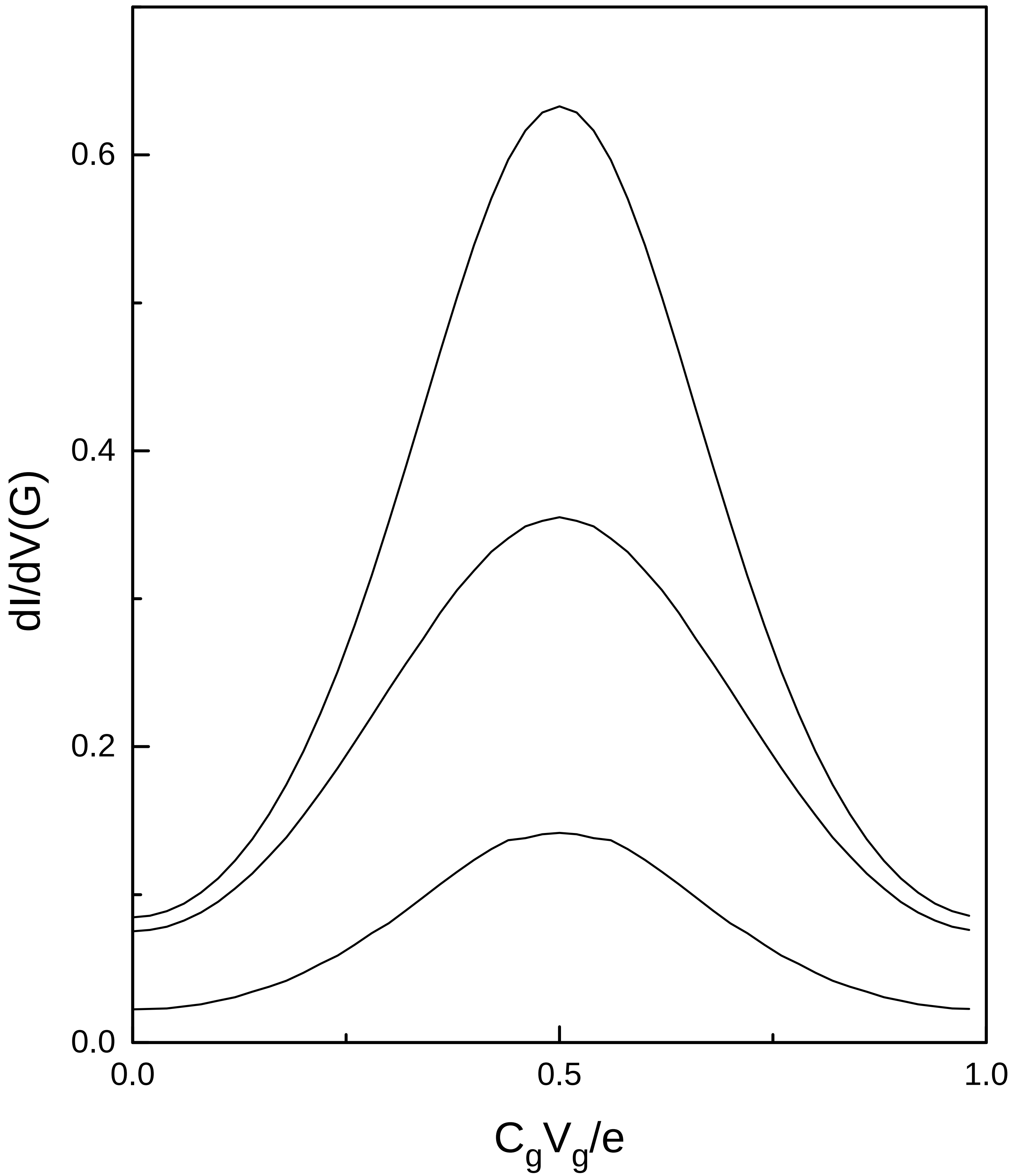
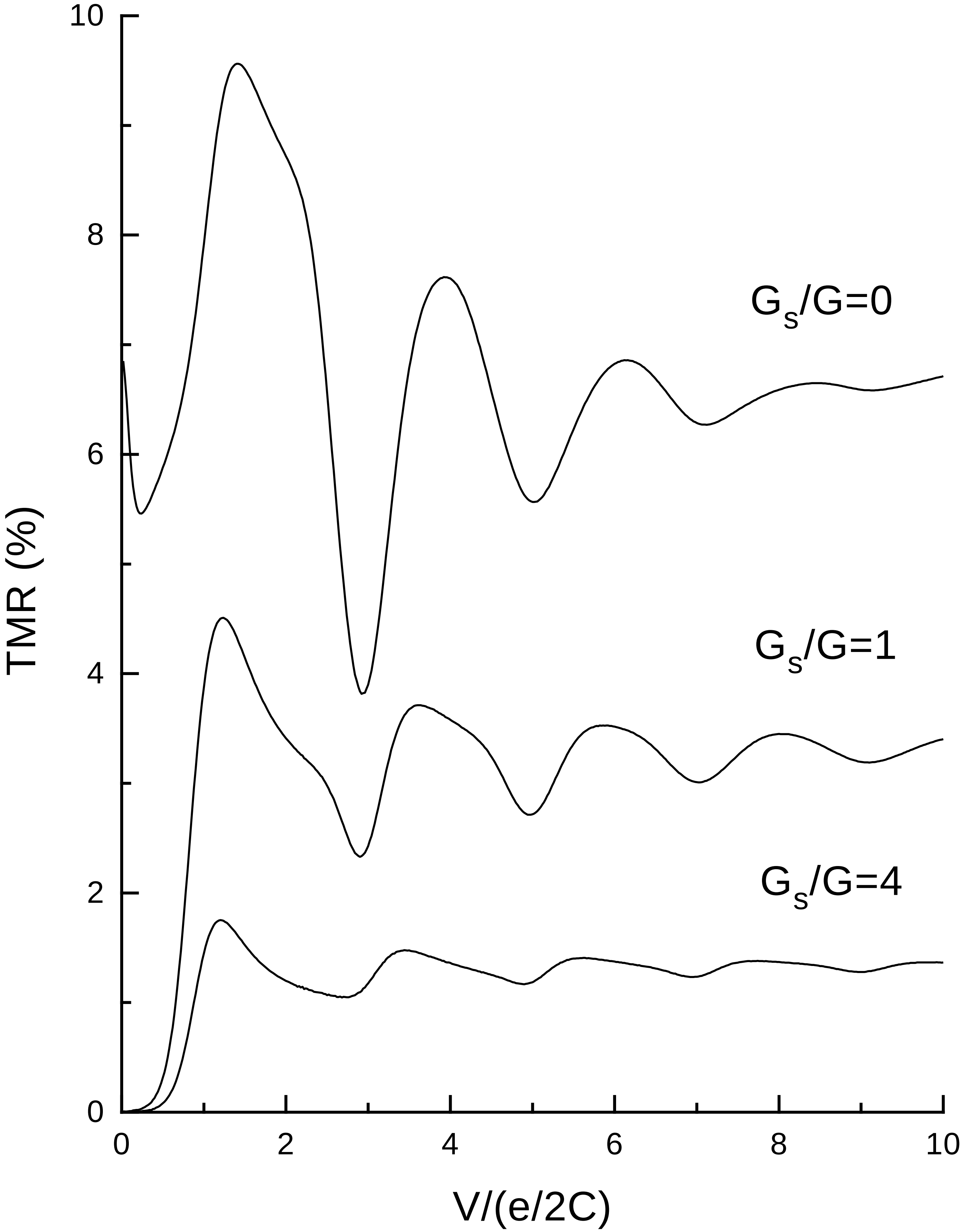
C 1 /2
C 2 /2

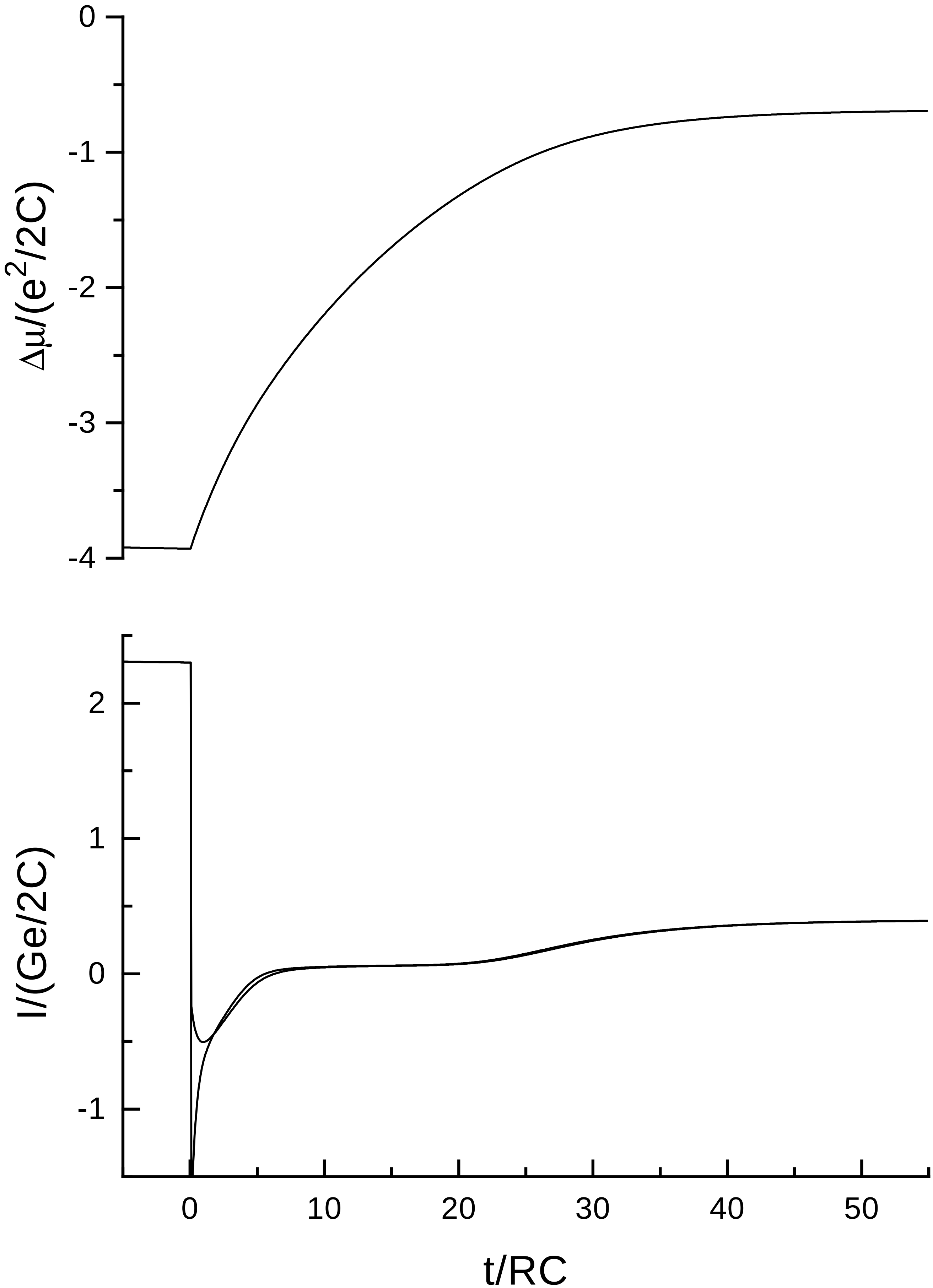

2.0
1.5
1.0
0.5
0.0
I(Ge/2C)
0
1
2
3
4
5
6
eV/(e^2/2C)



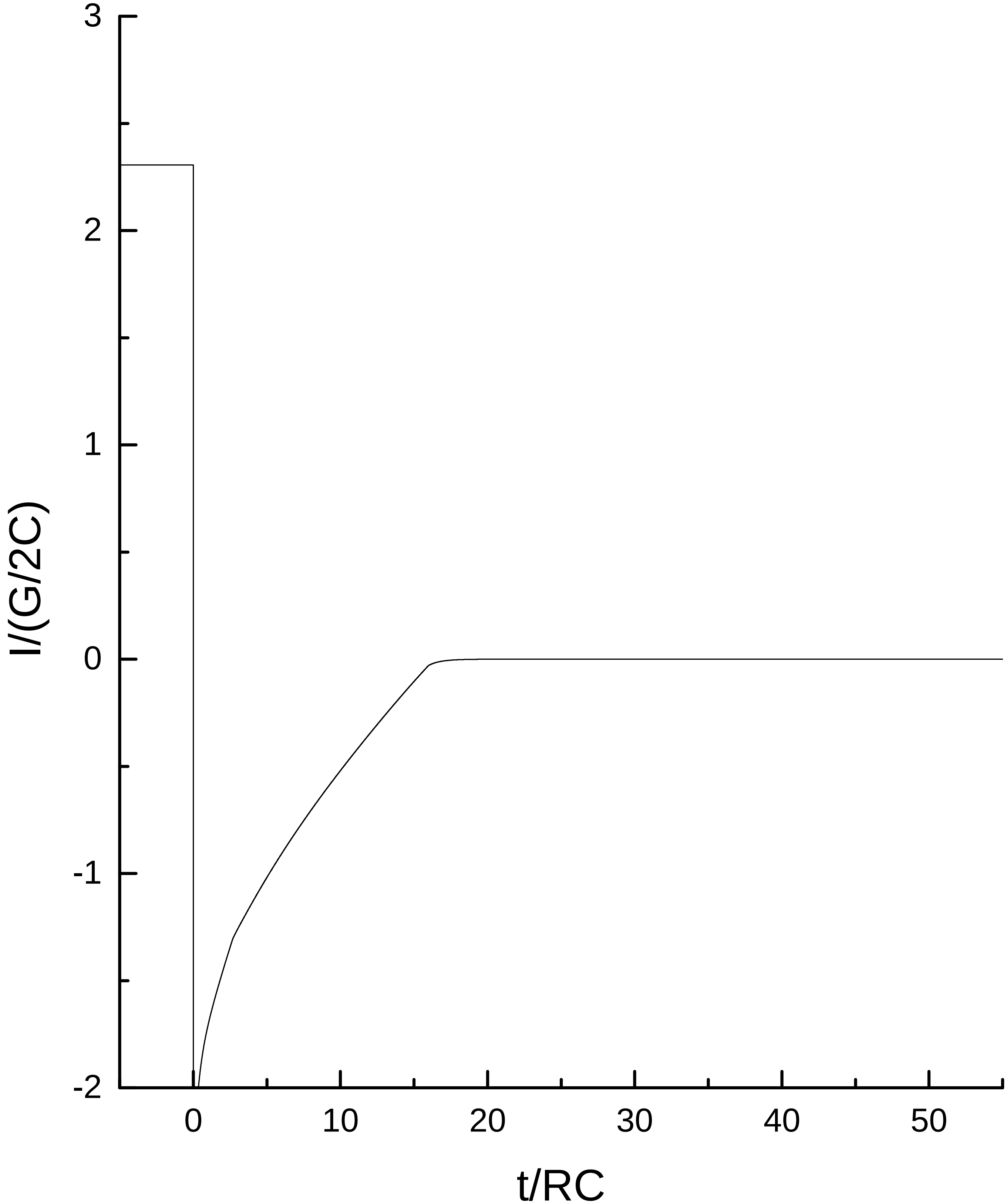
0.6
0.4
0.2
0.0
0.0
0.5
1.0
dI/dV(G)
C_gV_g/e



10
8
6
4
2
0
TMR (%)
0
2
4
6
8
10
V/(e/2C)
G_S/G=0
G_S/G=1
G_S/G=4



0
-1
-2
-3
-4
Δμ/(e²/2C)
2
1
0
-1
I/(Ge/2C)
0
10
20
30
40
50
t/RC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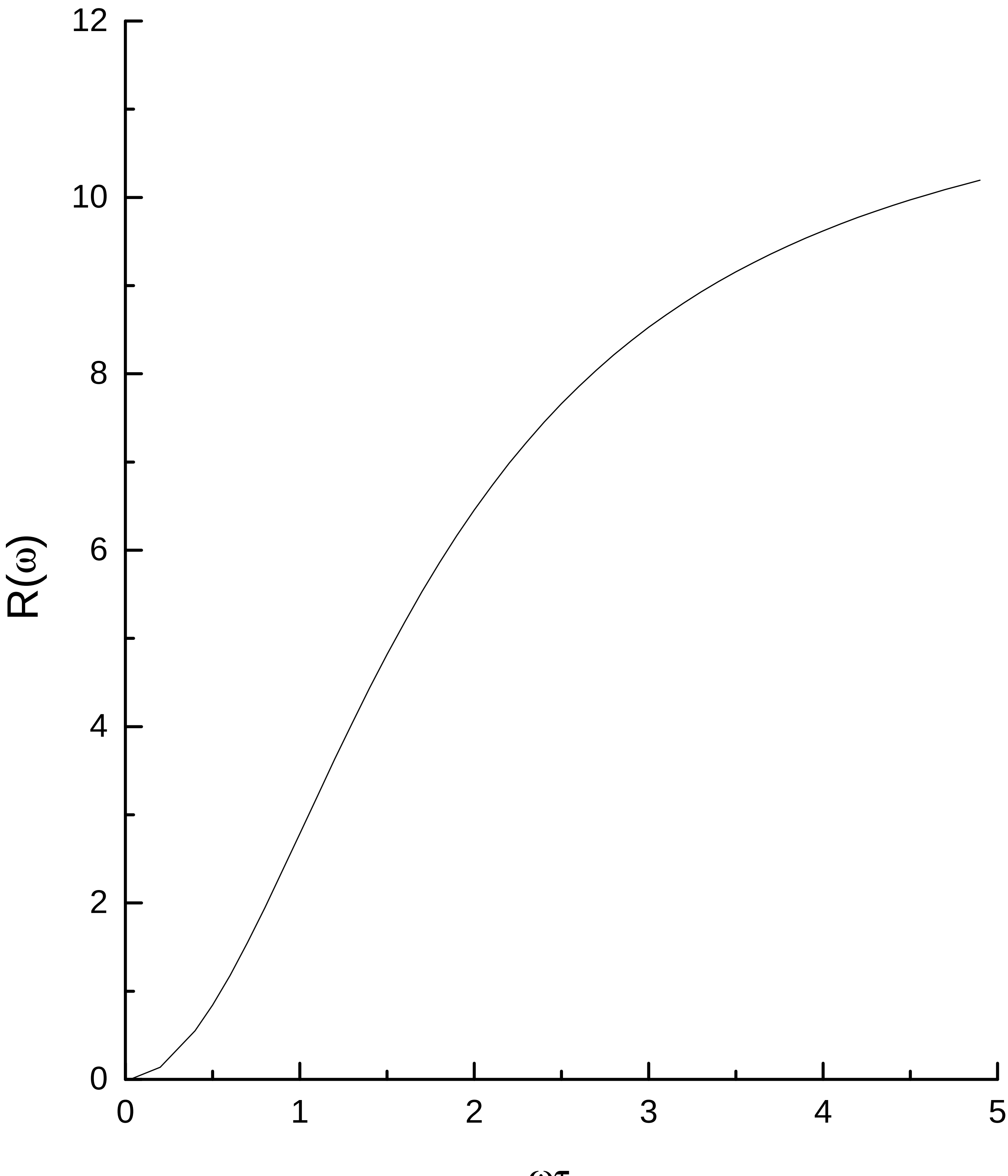
2
1
0
-1
-2
I/(G/2C)
0
10
20
30
40
50
t/RC



12
10
8
6
4
2
0
R(ω)
0
1
2
3
4
5
ωτsf